Slow dynamics and aging in glassy ferrofluids

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For long colloidal dispersions have been considered as model systems to mimic the glass transition of molecular materials, both experimentally and theoretically. For colloidal particles interacting with a repulsive interparticle potential (either short range with hard sphere repulsion, or longer range with screened coulomb repulsion for charge-stabilized particles), a generic behavior near the glass transition is the existence of two distinct relaxation processes : a fast β-process due to the rattling of the spheres inside the cage made by the neighboring particles and a much slower process named α-relaxation.
We present here novel experimental results where the α-relaxation was investigated in details for a strongly repulsive glassy colloidal dispersion. The system consists on aqueous dispersions of magnetic nanoparticles, bearing a superficial charge density, with tunable interparticle interaction via the ionic strength of the system and its osmotic pressure. An anisotropic contribution to the interparticle potential can be introduced in the system by applying an external magnetic field. We investigate its dynamic behavior (first without the presence of a magnetic field) with X-ray Photon Correlation Spectroscopy, at lengthscales close to interparticle distances. The large wavevector accessible with XPCS (together with the absence of multiple scattering in the X-ray regime [1]) make this study among the first to investigate the glassy dynamics of highly repulsive spherical nanoparticles on nanometric lengthscales.\\
These disordered systems are in a meta-stable state far from equilibrium and they relax very slowly. As a result, the correlation functions have an unusual behavior far from what is known for systems in equilibrium. If the temporal intensity autocorrelation function is measured over a relatively short period of time, it reflects the quasi-equilibrium properties over the measurement period and hence characterizes the system at a certain age. However, the correlation functions can vary substantially if they are measured at a different age, as the material will have relaxed, or aged, into a new metastable state. In this case, time-resolved 2D-XPCS allows the dispersion relations of magnetic nanoparticles in a glassy state to be probed; i.e. the age- and wavevector-dependence of the α-relaxation [2]. With an appropriate time-resolution, one can even characterize the dynamical fluctuations resulting from intermittency using X-ray Time Resolved Correlation (XTRC) statistical analysis [3]. This allows comparing simulation results, i.e the dynamical susceptibility or four-point susceptibility of such glassy systems.
The characteristic relaxation time, obtained as a fit to $g^2(Q,\tau)$ with compressed exponential, is observed to scale as Q^{-1} and increases with increasing age. Two aging regimes can then be clearly identified and present similarities with previously observed glassy aging-dynamics but in attractive glasses. Preliminary results with magnetic field will be presented.

[1] A. Robert, J. Wagner, T. Autenrieth, W. Härtl and G. Grübel, *J. Magn. Magn. Mat.* **289**, 47 (2005)
[2] A. Robert, E. Wandersman, E. Dubois, V. Dupuis and R. Perzynski, *Europhys. Lett.* **75**, 764 (2006)
[3] A. Duri and L. Cipelletti, *Europhys. Lett*. **76**, 972 (2006)